# **Supporting Information**

### *In situ* Scattering of Single Gold Nanorod Coupling with Monolayer

## **Transition Metal Dichalcogenides**

Aiqin Hu<sup>a,b</sup>, Weidong Zhang<sup>a,b</sup>, Shuai Liu<sup>a</sup>, Te Wen<sup>a</sup>, Jingyi Zhao<sup>a</sup>, Qihuang Gong<sup>a, b</sup>, Yu Ye<sup>a</sup>, and Guowei Lu<sup>a, b, \*</sup>

<sup>a</sup> State Key Laboratory for Mesoscopic Physics, Collaborative Innovation Center of Quantum Matter, Frontiers Science Center for Nano-optoelectronics, School of Physics, Peking University, Beijing 100871, China

<sup>b</sup> Collaborative Innovation Center of Extreme Optics, Shanxi University, Taiyuan, Shanxi 030006, China

Email: guowei.lu@pku.edu.cn.



Fig. S1. (a) Experiment setup and (b) the AFM topographic images during nanomanipulation demonstrate a single GNR was moved from the glass onto the TMDCs monolayer, and the scale bar is 500 nm.



Fig. S2. (a) Schematic of the sample, top view. (b) Scattering spectra of the pristine TMDCs (red) and TMDCs edge (black) without GNR. Comparing to the scattering intensity of the single GNR (blue), the scattering signal of the pristine TMDCs (red) and TMDCs edge (black) without GNR can be ignored. (c) PL spectra of pristine WS<sub>2</sub> (green),  $MoS_2$  (orange), WSe<sub>2</sub> (purple) and  $MoSe_2$  (pink) on glass substrate. The continuous-wave laser at a wavelength of 532 nm was employed as the illumination source. An oil-immersion objective lens with a numerical aperture (1.3) was used for both excitation and collection. And the diameter size of the laser focus spot was about ~500 nm.



Fig. S3. Scattering spectra of GNR before (black) and after (red) coupling to TMDCs, WS<sub>2</sub> (a), MoS<sub>2</sub> (b), WSe<sub>2</sub> (c), and MoSe<sub>2</sub> (d), respectively. The scattering spectra of GNR on the glass substrate is fitted by the Lorentz model (blue), and the scattering spectra of GNR-TMDCs hybrid is fitted by the Fano model (green). Coupling effect on scattering spectra between single GNR and WS<sub>2</sub>, redshift 14 nm (a), single GNR and MoS<sub>2</sub>, redshift 25 nm (b), single GNR and WSe<sub>2</sub>, redshift 18 nm (c), single GNR and MoSe<sub>2</sub>, redshift 22 nm (d).



Fig. S4. Scattering spectra of different GNRs coupling to WS<sub>2</sub> via using the AFM nanomanipulation technique. Scattering spectra of GNR on glass (black) and GNR on WS<sub>2</sub> (red).



Fig. S5. Scattering spectra of different GNRs coupling to MoS<sub>2</sub> via using the AFM nanomanipulation technique. Scattering spectra of GNR on glass (black) and GNR on MoS<sub>2</sub> (red).



Fig. S6. Scattering spectra of different GNRs coupling to WSe<sub>2</sub> via using the AFM nanomanipulation technique. Scattering spectra of GNR on glass (black) and GNR on WSe<sub>2</sub> (red).

As we all know, the strong coupling occurs when the energy exchange between a quantum emitter (TMDCs) and optical cavity (GNRs) is fast enough to overcome their respective dissipation rates. At first, we have analyzed our experiment data. And the results indicated that the Rabi splitting is not large enough when the detuning between plasmon and exciton energy is near zero. So it doesn't rigorously satisfy the criterion for strong coupling. For example, in fig. S6d, the resonant wavelength of the GNR is ~724 nm on the glass substrate. Considering of the redshift ~18 ± 2 nm (from fig. S7) when the GNR was moved from the glass substrate onto the WSe2 monolayer, the detuning between the plasmon and exciton energy is near to zero. And the hybrid's scattering peak splitting  $\Omega = 100 \text{ meV}$ , the linewidth of GNR's scattering spectrum  $\Gamma = 194 \text{ meV}$  (the GNRs employed in this experiment have a larger size, about 110nm×50nm, so they have broader linewidth), the linewidth of WSe2's PL spectrum  $\Gamma_{exciton} = 45 \text{ meV}$  (from fig. S2). Therefore, the results don't rigorously satisfy the criterion ( $\Omega > \frac{\Gamma + \Gamma_{exciton}}{\Gamma + \Gamma_{exciton}}$ 

2 ) for strong coupling.<sup>1-5</sup> So we conclude that the interaction is near strong coupling regime but not up to strong coupling regime.



Fig. S7. Scattering spectra of different GNRs coupling to MoSe<sub>2</sub> via using the AFM nanomanipulation technique. Scattering spectra of GNR on glass (black) and GNR on MoSe<sub>2</sub> (red).



Fig. S8. Redshifts of the scattering peak when the different GNRs (ten to twenty) were moved from the glass to the TMDCs (blue dots for WS<sub>2</sub>, red dots for MoS<sub>2</sub>, purple dots for WSe<sub>2</sub>, and green dots for MoSe<sub>2</sub>) monolayer.

### Numerical simulation

The finite-difference time-domain (FDTD) method, a powerful technique for metallic nanostructures with arbitrary geometries, is employed to calculate the optical responses. The individual gold nanorod is modeled as a cylinder capped with hemispheres at each end that was placed on a TMDCs layer on top of a 500-nm-thick SiO<sub>2</sub> layer. In addition, the nanorods with diameter ~ 50 nm and length ~120 ± 10 nm are employed in the simulations. The Drude–Lorentz dispersion model is used for the optical dielectrics of gold and the refractive indexes of the dielectric media are set to 1.49 for silica, 1.0 for air, 1.33 for water, and 1.47 for glycerol. TMDC dielectric functions contain real and imaginary parts.<sup>6</sup> And the thickness of the monolayer TMDCs is set to 1nm. The mesh grid is set 0.2 nm for TMDCs region and 1nm for other regions.



Fig. S9. Calculated the Far-field pattern of the GNR on the glass without and with the TMDCs in the upper and the lower half-spaces using the FDTD method. (a) and (d) Schematic of the GNR on the bare glass and the GNR on the TMDCs monolayer, respectively. (b) and (e) Backward far-field pattern of GNR without and with the TMDCs, respectively. (c) and (f) Forward far-field pattern of GNR without and with the TMDCs, respectively.

Considering the modification of the scattering directivity, we have calculated the far-field pattern of the GNR on the glass without and with the TMDCs in the upper and the lower half-spaces using the FDTD method. The intensity ratio of GNR without and with the TMDCs was calculated by the relation,

$$r_1 = \frac{\Sigma(I_{Back-glass})}{\Sigma(I_{For-glass})} = 2.1053 \quad r_2 = \frac{\Sigma(I_{Back-WSe2})}{\Sigma(I_{For-WSe2})} = 2.2379 \quad \text{, respectively. The presence of TMDCs with}$$

high refractive indexes make the ratio between the scattered signal in the lower and the upper half-spaces

$$\frac{r_2 - r_1}{r_1} = 0.063$$

enhance about ~6.3%, which arises from



Fig. S10. (a) Distance dependence of the GNR-MoSe<sub>2</sub> scattering spectra. The minus distance defines the position of GNR on glass, and the plus distance represents the position of GNR on TMDCs. The zero position represents the boundary of the TMDCs. (b) The AFM topographic images during nanomanipulation corresponding to scattering spectra in (a). The positions 1, 2, 3, 4, 5, 6 and 7 indicated the distance -0.43  $\mu$ m, -0.29  $\mu$ m, 0  $\mu$ m, 0.84  $\mu$ m, 1.01  $\mu$ m, -1.10  $\mu$ m and 1.21  $\mu$ m, respectively.



Fig. S11. (a) Distance dependence of the GNR-WSe<sub>2</sub> scattering spectra. (b) Scattering intensity (blue) and scattering fitting line (purple) changes during the nanomanipulation. The minus distance defines the position of GNR on glass, and the plus distance represents the position of GNR on TMDCs. The zero position represents the boundary of the TMDCs.



Fig. S12. (a) Distance dependence of the GNR- $WSe_2$  scattering spectra. (b) Scattering intensity changes during the nanomanipulation. The minus distance defines the position of GNR on glass, and the plus distance represents the position of GNR on TMDCs. The zero position represents the boundary of the TMDCs.



Fig. S13. Scattering spectra under different excitation polarization angles. (a) Scattering spectra and (b) scattering intensity of the GNR on the glass dependent on the excitation polarization. (b), (e) Scattering spectra and (d), (f) scattering intensity of the GNR on WSe<sub>2</sub> monolayer dependent with the excitation polarization. HEB, high-energy branch, the peak at the short wavelength. LEB, Low-energy branch, the peak at the long wavelength.



Fig. S14. Calculated and experimental scattering spectra dependent on the orientation of the GNR on TMDCs monolayer. (a) Calculated scattering spectra of GNR on  $WSe_2$  using the FDTD method. (b), (c) Experimental scattering spectra of GNR with different orientations on  $WSe_2$  and  $MoS_2$  monolayer, respectively. In the inset of (b) and (c), the AFM (1), (2) and (3) correspond to orientations 1, 2 and 3 in the experimental data.



Fig. S15. Scattering spectra of a single GNR on the monolayer WSe<sub>2</sub> in the air (black), water (red), and glycerol (blue). The LSP resonance of GNR at 653 nm (a), 664nm (b), and 683 nm (c) on glass are shown by purple dot line.



Fig. S16. Scattering spectra of a single GNR on a glass substrate without (a) and with the monolayer  $WSe_2$  (b) measured in air (red), water (blue), and glycerol (green). (c) Linear fitting the scattering intensity with slope ~ - 1.84 and ~-1.94, respectively.

### Line-shape fitting of the experimental spectra with the Fano model

The scattering cross-section spectra  $\sigma_{sc}(\omega)$  of a nanostructure supporting dark and bright modes can be expressed in the following form<sup>7</sup>

$$\sigma_{sc}(\omega) = \sigma_{ex}(\omega)\sigma_{pl}(\omega) \tag{1}$$

$$\sigma_{sc}(\omega) = \frac{\left(\frac{\omega^2 - \omega_{ex}^2}{2W_{ex}\omega_{ex}} + q\right)^2 + b}{\left(\frac{\omega^2 - \omega_{ex}^2}{2W_{ex}\omega_{ex}}\right) + 1}, \quad \sigma_{pl}(\omega) = \frac{a^2}{\left(\frac{\omega^2 - \omega_{pl}^2}{2W_{pl}\omega_{pl}}\right) + 1}$$
(2)

Here,  $\omega_{ex}$  and  $W_{ex}$  are the resonant frequency and the half-width of resonance line at half-maximum of the exciton.  $\omega_{pl}$  and  $W_{pl}$  are the resonant frequency and the half-width of resonance line at half-maximum of the plasmonic resonances. q is the Fano asymmetry parameter, b is the damping parameter originating from intrinsic losses, a is the maximal amplitude of the resonance.



Fig. S17. Scattering spectra (gray line) of GNR-WSe<sub>2</sub> monolayer fitted by Fano lineshape (green line). The curves 1 (blue dashed ones) correspond to Eqs. (1) and (2), whereas the curves 2 (yellow dashed ones) are the additional Lorentzian signals, which are caused by other surface plasmon resonances.<sup>7</sup>



Fig. S18. Scattering spectra of GNR-WSe<sub>2</sub> monolayer in the air (black), water(red), and oil (blue) fitted by Fano lineshape (purple line).

The "q" is the Fano asymmetry parameter. Commonly, the SPs have much stronger dipole moment compared to excitons in TMDC monolayers, which causes  $|q| \ll 1$  in a plasmon–exciton system and the asymmetrical Fano lineshapes are indistinguishable. When the dipole moment of the exciton subsystem (which depends on the number of excitons) becomes comparable to that of SPs, i.e.,  $|q| \approx 1$ , more distinct asymmetrical Fano lineshapes appear. <sup>7-8</sup> Firstly, the detuning between the plasmon and exciton energy is more significant when the GNR-WSe2 in air. So the  $|q| \ll 1$  because the plasmon-exciton interaction is weaker. Then the detuning between the plasmon and exciton energy is constant of the solvent increase. So the plasmon-exciton interaction becomes stronger and the |q| is near to 1.

### **Classical oscillator modeling**

In general, for an classical oscillator (  $\ddot{mx} = -\gamma \dot{x} - kx - eE_0 e^{-i\omega_{ex}t}$ )<sup>9</sup>, the light scattering can be written as

$$I_{Sca}(\omega) \sim \frac{2\pi A^2 \omega^2}{|\omega^2 - \omega_c^2 - 2i\beta\omega|^2}, \text{ where } \frac{k}{m} = \omega_0^2, \frac{\gamma}{m} = 2\beta, \frac{eE_0}{m} = A, \omega_c^2 = \omega_0^2 - \beta^2, (\omega_c \sim \omega_0, \text{ if } \omega_0^2 \gg \beta^2). x \text{ is the}$$

displacement of the electron,  $\gamma$  is the damping rate,  $E_0$  and  $\omega_{ex}$  are the electric field and resonant frequency of the light wave.  $\omega_0$  is the natural resonant frequency,  $\beta$  originates from intrinsic losses, A is the maximal amplitude of the resonance. When the loss ( $\beta$ ) of the oscillator system decreases, the linewidth of scattering narrowed and the intensity increases, shown in the main text part (Fig. 5c).

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